PATENT

Docket No.: CL/V-32421A

## **CLAIM AMENDMENTS**

Please amend claims 1, 3, and 7-8 as follows:

- 1. (Currently amended) A polymerizable material for making an ophthalmic device, comprising:
  - (a) a water-soluble polyvinyl alcohol having crosslinkable groups; and
  - (b) one or more members selected a modifier which is different from but miscible with the water-soluble polyvinyl alcohol and presented in the polymerizable material in an amount sufficient to improve one or more physical proporties of the ophthalmic device made from the polymerizable material, wherein the one or more physical proporties are selected from the group consisting of stress at break (N/mm²), percentage of clongation at break, toughness or energy to break (N-mm), and susceptibility to fracture, wherein the modifier is-selected-from the group consisting of nanoparticles having a hydrophilic surface, a copolymer composed of hydrophilic vinylic monomer units and hydrophobic vinylic monomer units, a non-crosslinkable polyurethane, a crosslinkable polyurethane with vinyl groups, and mixtures thereof, wherein the copolymer is a copolymerization product of a mixture including at least one-hydrophilic vinylic monomer units are derived from the group consisting of hydroxy-substituted alkylmethacrylate, hydroxy-substituted alkylacrylate, N-vinyl-lactams, N,N-dialkyl-methacrylamide, and a mixture thereof and the at least one hydrophobic vinylic monomer units are derived from the group consisting of a C<sub>1</sub>-C<sub>18</sub>-alkylacrylate, a C<sub>1</sub>-C<sub>18</sub>-alkylmethacrylates, a di-C<sub>1</sub>-C<sub>7</sub> alkylamino-C<sub>1</sub>-C<sub>7</sub> alkylacrylate, an acrylonitrile, a methacrylonitrile, a vinyl-C<sub>1</sub>-C<sub>18</sub>-alkanoate, a C<sub>2</sub>-C<sub>18</sub>alkenes, a C<sub>2</sub>-C<sub>18</sub>-halo-alkenes, styrene, a C<sub>1</sub>-C<sub>6</sub>-alkylstyrene, a vinylalkylether in which the alkyl moiety has 1 to 6 carbon atoms, a C<sub>2</sub>-C<sub>10</sub>-perfluoralkylacrylate, a C<sub>2</sub>-C<sub>10</sub>perfluoralkylmethacrylates, an acryloxyalkylsiloxane, a methacryloxy-alkylsiloxane, glycidyl methacrylate, butoxyethylacrylate, a mixture thereof, wherein the percentage of the hydrophobic vinylic monomer units of the copolymer is sufficient high to impart at least one desired physical property to said ophthalmic device, wherein the percentage of the <del>at least one</del> hydrophilic monomer <u>units of the copolymer</u> is <del>presented in the mixture</del> sufficient high to render the resultant-copolymer miscible with the water-soluble polyvinyl alcohol,

wherein component (b) is presented in the polymerizable material in an amount sufficient to improve one or more physical properties of the ophthalmic device made from the

polymerizable material, wherein the one or more physical properties are selected from the group consisting of stress at break (N/mm2), percentage of elongation at break, toughness or energy to break (N\pimm), and susceptibility to fracture.

- 2. (Canceled).
- 3. (Currently amended) A polymerizable material of claim 3₂, wherein said water-soluble polyvinyl alcohol is a polyhydroxyl compound which has a weight average molecular weight of at least about 2000 and which comprises from about 0.5 to about 80%, based on the number of hydroxyl groups in the poly(vinyl alcohol), of units of the formula I, I and II, I and III, or I and II and III

$$\begin{array}{c|c}
 & H_2 & H_2 \\
 & C & C \\
 & C & C \\
 & R_3 & O \\
 & R_1 & R_2
\end{array}$$

in which R is alkylene having up to 12 carbon atoms,  $R_1$  is hydrogen or lower alkyl,  $R_2$  is an olefinically unsaturated, electron-withdrawing, crosslinkable radical having up to 25 carbon atoms, and  $R_3$  is hydrogen, a  $C_1$  - $C_6$  alkyl group or a cycloalkyl group,

$$\begin{array}{c|c}
 & H_2 & H_2 \\
 & C & C \\
 & CH & CH \\
 & R_3 & O \\
 & O & O \\
 & R & R_7
\end{array}$$
II

wherein R and R<sub>3</sub> are as defined above, and R<sub>7</sub> is a primary, secondary or tertiary amino group or a quaternary amino group of the formula  $N^{+}(R')_3X^{-}$ , in which each R', independently of the others, is hydrogen or a C<sub>1</sub> -C<sub>4</sub> alkyl radical and X is HSO<sub>4</sub>, F<sup>-</sup>, Cl<sup>-</sup>, Br<sup>-</sup>, I<sup>-</sup>, CH<sub>3</sub> COO<sup>-</sup>, OH<sup>-</sup>, BF<sup>-</sup>, or H<sub>2</sub>PO<sub>4</sub><sup>-</sup>,

$$\begin{array}{c|c}
 & H_2 & H_2 \\
 & C & C \\
 & R_3 & O \\
 & & R_3 & O
\end{array}$$
III

in which R and  $R_3$  are as defined above, and  $R_8$  is the radical of a monobasic, dibasic or tribasic, saturated or unsaturated, aliphatic or aromatic organic acid or sulfonic acid.

4. (Original) A polymerizable material of claim 3, wherein said water-soluble polyvinyl alcohol is a polyhydroxyl compound which has a molecular weight of at least about 2000 and which comprises from about 0.5 to about 80%, based on the number of hydroxyl groups in the poly(vinyl alcohol), of units of the formula I, wherein R<sub>2</sub> is a radical of formula IV or formula V

$$-CO-NH-(R_5-NH-CO-O)_0 -R_6 -O-CO-R_4$$
 (IV)

$$-[CO-NH-(R_5-NH-CO-O)_q -R_6 -O]_p-CO-R_4$$
 (V)

in which p and q, independently of one another, are zero or one, and  $R_5$  and  $R_6$ , independently of one another, are lower alkylene having 2 to 8 carbon atoms, arylene having 6 to 12 carbon atoms, a saturated bivalent cycloaliphatic group having 6 to 10 carbon atoms, arylenealkylene or alkylenearylene having 7 to 14 carbon atoms or arylenealkylenearylene having 13 to 16 carbon atoms, and in which  $R_4$  is an olefinically unsaturated copolymerizable radical having 2 to 24 carbone atoms, preferably having 2 to 8 carbonatoms, more preferably having 2 to 4 carbon atoms.

- 5. (Withdrawn) A polymerizable material of claim 3, wherein said modifier is composed of the nanopaticles having a hydrophilic surface.
- (Withdrawn) A polymerizable material of claim 5, wherein the nanoparticles are nanosized silica fillers.
- 7. (Currently amended) A polymerizable material of claim 3, wherein said <u>component (b)</u> <u>comprises a modifier is composed of one or more</u> copolymers <u>each having hydrophobic</u> groups or units for imparting at least one desired physical property to said ophthalmic device and hydrophilic groups or units in an amount-sufficient to render the copolymer miscible with the crosslinkable polyvinyl alcohol.
- (Currently amended) A polymerizable material of claim 7, wherein the hydrophilic monomer units of said copolymer modifier is a N-vinyl lactam copolymer which is a

copolymerization product of at least one N-vinyl lactam with one or more hydrophobic monomer, wherein said at least one N-vinyl lactam having has a structure of formula (VI)

$$R_{20}$$
 $R_{19}$ 
 $R_{21}$ 
 $R_{19}$ 
 $R_{19}$ 
 $R_{21}$ 
 $R_{19}$ 
 $R_{19}$ 
 $R_{21}$ 
 $R$ 

in which  $R_{19}$  is an alkylene di-radical having from 2 to 8 carbon atoms,  $R_{20}$  is hydrogen,  $C_1$ - $C_7$  alkyl, aryl having up to 10 carbon atoms, aralkyl or alkaryl having up to 14 carbon atoms, and  $R_{21}$  is hydrogen or lower alkyl having up to 7 carbon atoms.

- 9. (Original) A polymerizable material of claim 8, wherein said N-vinyl lactam is N-vinyl pyrrolidone.
- 10. (Withdrawn) A polymerizable material of claim 7, wherein said modifier is a N,N-dialkylmethacrylamide copolymer which is a copolymerization product of a N,N-di-C<sub>2</sub>-C<sub>4</sub> alkyl methacrylamide with at least one hydrophobic monomer.
- 11. (Withdrawn) A polymerizable material of claim 10, wherein the N,N-di-C<sub>2</sub>-C<sub>4</sub> alkyl methacrylamide is N,N-dimethylmethacrylamide.
- 12. (Withdrawn) A polymerizable material of claim 7, wherein said modifier is a non-crosslinkable polyurethane having a molecular weight of at least about 2000, or a crosslinkable polyurethane.
- 13. (Withdrawn) A polymerizable material of claim 12, wherein said non-crosslinkable polyurethane is the reaction product of an isocyanate-capped polyurethane with water and amine, wherein said crosslinkable polyurethane is the reaction product of the isocyanate-capped polyurethane with an ethylenically unsaturated amine (primary or secondary amine) or an ethylenically unsaturated monohydroxy compound, wherein said isocyanate-capped polyurethane is a copolymerization product of
  - (a) at least one polyalkylene glycol of formula

$$HO-(R_{9}-O)_{n}-(R_{10}-O)_{m}-(R_{11}-O)_{l}-H$$
 (1)

wherein  $R_9$ ,  $R_{10}$ , and  $R_{11}$ , independently of one other, are each linear or branched  $C_2$ - $C_4$ -alkylene, and n, m and l, independently of one another, are each a number from 0 to 100, wherein the sum of (n+m+l) is 5 to 100,

- (b) at least one branching agent selected from the group consisting of
  - (i) a linear or branched aliphatic polyhydroxy compound of formula

PATENT

Docket No.: CL/V-32421A

$$R_{12}\text{-}(OH)_X \tag{2},$$

wherein  $R_{12}$  is a linear or branched  $C_3$ - $C_{18}$  aliphatic multi-valent radical and x is a number  $\geq 3$ ,

- (ii) a polyether polyol, which is the polymerization product of a compound of formula (2) and a glycol,
- (iii) a polyester polyol, which is the polymerization product of a compound of formula (2), a dicarboxylic acid or a derivative thereof and a diol, and
- (iv) a cycloaliphatic polyol selected from the group consisting of a C5-C8-cycloalkane which is substituted by  $\geq$  3 hydroxy groups and which is unsubstituted by alkyl radical, a C5-C8-cycloalkane which is substituted by  $\geq$  3 hydroxy groups and which is substituted by one ore more C<sub>1</sub>-C<sub>4</sub> alkyl radicals, and an unsubstituted monoand disaccharide,
- (v) an aralkyl polyol having at least three hydroxy  $C_1$ - $C_4$  alkyl radicals, and (c) at least one di- or polyisocyanate of formula

$$R_{13}\text{-}(NCO)_{V} \tag{3}$$

wherein  $R_{13}$  the multivalent radical of a linear or branched  $C_3$ - $C_{24}$  aliphatic polyisocyanate, the multivalent radical of a  $C_3$ - $C_{24}$  cycloaliphatic or aliphatic-cycloaliphatic polyisocyanate, or the multivalent radical of a  $C_3$ - $C_{24}$  aromatic or araliphatic polyisocyanate, and y is a number from 2 to 6,

wherein said ethylenically unsaturated monohydroxy compound is a hydroxy-substituted lower alkylacrylate, a hydroxy-substituted lower alkylmethacrylate, a hydroxy-substituted lower alkyl-acrylamides, a hydroxy-substituted lower alkyl-methacrylamide, or a hydroxy-substituted lower alkylvinylether, wherein said ethylenically unsaturated amine has formula (4), (4') or (4")

$$R_{14} \longrightarrow \stackrel{H}{N} \longrightarrow \stackrel{O}{C} \longrightarrow \stackrel{O}{\downarrow}_{i} \longrightarrow$$

PATENT

Docket No.: CL/V-32421A

$$R_{15}$$
 $R_{15}$ 
 $CH_2$ 
 $CH_2$ 
 $CH_2$ 
 $CH_2$ 

In which, I, j and k, independent of one another, are o or 1;

 $R_{14}$  is hydrogen, a linear or branched  $C_1$ - $C_{24}$  alkyl, a  $C_2$ - $C_{24}$  alkoxyalkyl, a  $C_2$ - $C_{24}$  alkylcarbonyl, a  $C_2$ - $C_{24}$  alkoxycarbonyl, an unsubstituted or  $C_1$ - $C_4$  alkyl- or  $C_1$ - $C_4$  alkoxy-substituted  $C_6$ - $C_{10}$  aryl, a  $C_7$ - $C_{18}$  aralkyl, a  $C_{13}$ - $C_{22}$  arylalkylaryl, a  $C_3$ - $C_8$  cycloalkyl, a  $C_4$ - $C_{14}$  cycloalkylalkyl, a  $C_7$ - $C_{18}$  cycloalkylalkylcycloalkyl, a  $C_5$ - $C_{20}$  alkylcycloalkylalkyl, or an aliphatic-heterocyclic radical;

Z is a  $C_1$ - $C_{12}$  alkylene radical, phenylene radical or  $C_7$ - $C_{12}$  aralkylene radical;  $R_{15}$  and  $R_{15}$ , independently of each other, are hydrogen,  $C_1$ - $C_4$  alkyl or halogen; and

Q is a radical of formula (5)

$$--- (Z')_r --- C --- C --- C R_{16}$$

$$R_{18}$$

$$(5)$$

wherein r is the number 0 or 1,

each of  $R_{16}$  and  $R_{17}$  independently of the other is hydrogen,  $C_1$ - $C_4$  alkyl, phenyl, carboxy or halogen,

R<sub>18</sub> is hydrogen, C<sub>1</sub>-C<sub>4</sub> alkyl or halogen, and

Z' is a linear or branched  $C_1$ - $C_{12}$  alkylene, an unsubstituted phenylene, an  $C_1$ - $C_4$  alkyl- or  $C_1$ - $C_4$  alkoxy-substituted phenylene, or a  $C_7$ - $C_{12}$  aralkylene.

14. (Withdrawn) A polymerizable material of claim 13, wherein component (a) consists of one or more polyalkylene glycols of formula (1a)

$$HO-(CH_2-CH_2-O)_n-(CHY_1-CHY_2-O)_m-H$$
 (1a)

wherein one of radicals Y<sub>1</sub> and Y<sub>2</sub> signifies methyl and the other radical signifies hydrogen, and n and m, independently of one another, each denote a number from 0 to 50, wherein the sum of (n+m) is 8 to 50,

wherein component (b) consists of one or more linear or branched aliphatic polyhydroxy compounds of formula (2), in which x is a number from 3 to 8,

wherein component (c) consists of one or more diisocyanates of formula (3a)

$$OCN-R_5-NCO$$
 (3a)

wherein  $R_5$  is a linear or branched C3-C18-alkylene, an unsubstituted or C1-C4-alkylsubstituted or C1-C4-alkoxy-substituted C6-C10-arylene, a C7-C18-aralkylene, a C6-C10-arylene-C1-C2-alkylene-C6-C10-arylene, a C3-C8-cyclo-alkylene, a C3-C8-cycloalkylene-C1-C6-alkylene, a C3-C8-cycloalkylene-C1-C6-alkylene, a C3-C8-cycloalkylene-C1-C6-alkylene, or a C1-C6-alkylene-C3-C8-cycloalkylene-C1-C6-alkylene, wherein said ethylenically unsaturated amine is selected from the group consisting of mono-C1-C4 alkylamino-C1-C4 alkyl-acrylates, mono-C1-C4 alkylamino-C1-C4 alkyl-methacrylates, di- C1-C4 alkylamino- C1-C4 alkyl-acrylates and di- C1-C4 alkylamino- C1-C4 alkyl-methacrylates, and wherein said ethylenically unsaturated hydroxy compound is selected from the group consisting of hydroxy-substituted C1-C6 alkylmethacrylates.

15. (Withdrawn) A polymerizable material of claim 14, wherein said ethylenically unsaturated amine is 2-terbutylaminoethylmethacrylate or 2-terbutylaminoethylacrylate, wherein said ethylenically unsaturated hydroxy compound is 2-hydroxyethylmethacrylate or 2-hydroxyethylcrylate, wherein component (c) consists of a diisocyanate selected from the group consisting isophorone diisocyanate (IPDI), toluylene-2,4-diisocyanate (TDI), methylenebis(cyclohexyl-isocyanate), 1,6-diisocyanato-2,2,4-trimethyl-n-hexane (TMDI), methylenebis(phenyl-isocyanate) and hexamethylene-diisocyanate (HMDI).

16-46. (Canceled)